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# **The Next Nuclear Challenge- How do we Dispose of the Excess Nuclear Materials?**

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# The Next Nuclear Challenge - How Do We Dispose of the Excess Nuclear Materials?

by

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## ABSTRACT

The "Cold War" was not fought only by soldiers but by scientists and engineers in Laboratories and plants located throughout the world. With the fall of the Berlin Wall, the "Cold War" was effectively over, but the weapons of nuclear war remained. Following signing of START 2 (Strategic Arms Reduction Treaty) in 1993, up to 100 tonnes of weapons usable plutonium is expected to be declared excess by the Super Powers. Steps must be taken to address the proliferation risks associated with this plutonium. Again the scientist and engineers, who were the "Cold War" warriors, are being asked to develop methods to disposition this plutonium such that it can never again be used for weapons. Will we burn the plutonium in reactors or immobilize the plutonium either in a glass or ceramic matrix? Interesting challenges face chemists and chemical engineers developing immobilization techniques to render the plutonium both environmentally benign, and proliferation resistant.

## INTRODUCTION AND BACKGROUND

World War II effectively ended with the dropping of the plutonium bomb over Nagasaki, Japan. In the struggle for power and influence in the aftermath of WWII, the world entered into an arms race, primarily between the United States and the Soviet Union and their respective allies. This state of tension and military rivalry, known as the Cold War, lasted from about 1945 until about 1990.

This "Cold War" was not "fought" by soldiers alone on a battle line with guns and tanks – instead it was waged with many weapons: diplomatic, propaganda, economic, and military. The main propaganda and military weapons were nuclear. The nuclear part of the Cold War was waged by scientists, engineers, and technicians in the weapons laboratories and nuclear material production plants of the two superpowers:

- Nuclear weapons were designed and tested in laboratories such as the Lawrence Livermore National Laboratory and the Los Alamos National Laboratory in the United States; Arazamas-16 and Chelyabinsk-70 in the Soviet Union.

- Nuclear materials were produced in plants such as Savannah River and Hanford in the United States; Tomsk-7 and Chelyabinsk-65 (Mayak) in the Soviet Union.
- Nuclear weapons were shaped and assembled in plants such as Rock Flats and Pantex in the United States; Tomsk-7, Chelyabinsk-70 and Arzamas-16 in the Soviet Union.

Tearing down the Berlin Wall in 1989 signaled that the Cold War was at an end. This symbol preceded the reality, for the work of ending the Cold War—managing the legacy of the nuclear weapons-usable materials—remains yet to be done.

The governments of the United States and Russia have taken the first steps toward nuclear disarmament by negotiating the Strategic Arms Reduction Treaties (START I and START II). Under START I, which was ratified in 1991, both countries have agreed to reduce their nuclear weapons to approximately 6000 warheads, and have already begun to do so by dismantling weapons on the order of 1300 to 2000 each year. START II, when it is ratified, would reduce the numbers further to between 3000–3500.

The dismantling of nuclear weapons and ceasing to manufacture new weapons, while positive for world peace, has raised yet another problem: how do we manage the fissile materials recovered from the weapons or in inventories that would no longer be used in weapons manufacture? These materials—primarily plutonium and highly enriched uranium—pose environmental, safety, and health concerns and there is a serious risk of nuclear proliferation from the resulting growing stockpiles. Nuclear weapons or the fissile materials recovered from them could fall into the hands of terrorists or rogue, non-nuclear nations through theft or diversion. The U.S. National Academy of Sciences (NAS) report on the management and disposition of excess weapons plutonium characterized this as a “clear and present danger.” This nuclear danger is, in many ways, more diffuse, harder to manage, and more dangerous than the nuclear tensions of the Cold War era.

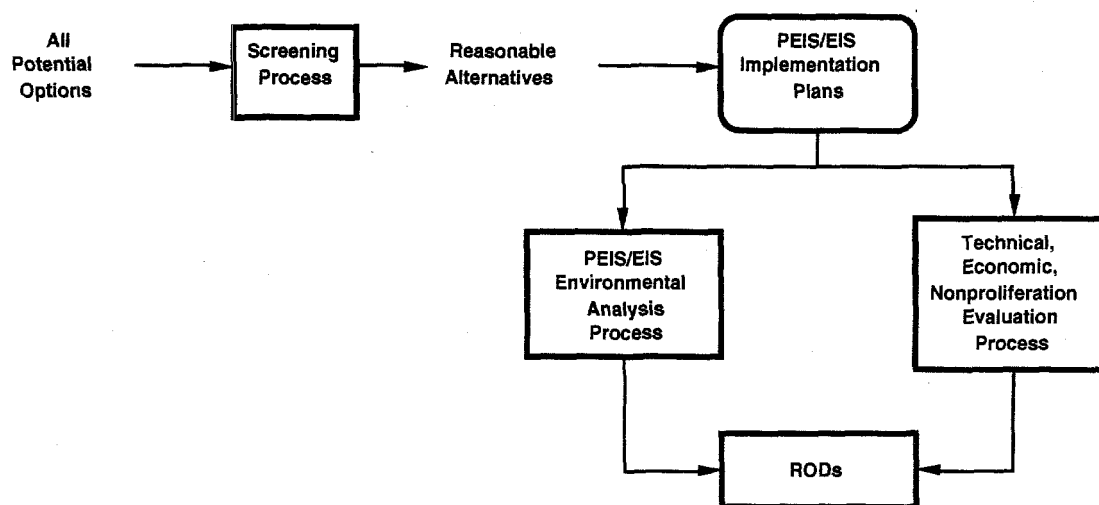
In September 1993, President Clinton issued the U.S. Nonproliferation and Export Control Policy in response to the growing threat of nuclear weapons proliferation. Further, in January 1994, President Clinton and Russia’s President Yeltsin issued a *Joint Statement Between the United States and Russia on Nonproliferation of Weapons of Mass Destruction and Means of Their Delivery*.

In the U.S., the Department of Energy (DOE) has the technical lead for the disposition studies, acting as a member of the Interagency Working Group of the White House Office of Science and Technology Policy. On January 24,

1994, a DOE-wide project for control and disposition of surplus fissile materials, which later became the Office of Fissile Materials Disposition (MD), was created. MD, through task teams composed of experts from national laboratories, production sites, universities, industry, and other DOE programs, e.g. Civilian Radioactive Waste Management, (RW), have completed a comprehensive review of long-term options for surplus fissile materials storage and disposition, taking into account technical, nonproliferation, environmental, budgetary, and economic considerations. In furthering this policy, DOE's objectives included:

1. Establishing transparent and irreversible nuclear arms reduction.
2. Strengthening national and international arms control efforts by providing an exemplary model for storage of all weapons-usable fissile materials and the disposition of surplus weapons-usable materials.
3. Ensuring that storage and disposition of weapons-usable fissile materials are carried out in compliance with ES&H standards.
4. Minimizing the prospect that surplus U. S. weapons-usable fissile materials could be re-introduced into the arsenals from which they came, therefore increasing the likelihood of reciprocal measures by Russia and other nuclear powers.
5. Minimizing the risk that surplus U. S. weapons-usable fissile materials could be obtained by unauthorized parties.
6. Accomplishing these objectives in a timely and cost-effective manner.

DOE announced that its evaluations leading to Records of Decision (ROD) would be carried out via the process depicted in Figure 1.



**Figure 1. Evaluation Process Leading To Records of Decision.**

DOE also announced that any decision to commence implementation of surplus weapons-usable fissile materials disposition would be made on a broad domestic and international context taking into account any arrangements or agreements reached with the Russian government. These decisions will involve other Executive branch agencies in addition to the Department of Energy.

Screening criteria were developed based on the policy objectives articulated in the President's Nonproliferation and Export Control Policy of September 1993 and the January 1994 *"Agreement between the United States and Russia on Nonproliferation of Weapons of Mass Destruction and their means of Delivery,"* as well as the analytical framework established by the NAS in their study on disposition of surplus plutonium. Before finalizing the original criteria, DOE obtained public input on screening criteria to be utilized and options to be evaluated as part of the public scoping process. As an initial step in the National Environmental Policy Act (NEPA) process and as announced in the Notice of Intent (NOI), a series of 12 Scoping Meetings were held at twelve locations across the country from August to October 1994. Members of the interested public were briefed by DOE on the overall long-term storage and disposition considerations, criteria, and evaluation process, including the screening process. During these meetings, questionnaires were provided for public input on (1) validity and relative importance of the criteria, and (2) additional criteria that should be considered. Responses received were evaluated, and modification to the criteria made and documented in the *"Summary Report of the Screen Process, DOE/MD-0002,* March 29, 1995. Based upon this additional input from the public the following set of criteria have been used for rating individual options:

1. Resistance to Theft and Diversion by Unauthorized Parties
2. Resistance to Retrieval, Extraction, and Reuse by the Host Nation
3. Technical Viability
4. Environmental, Safety, and Health
5. Cost Effectiveness
6. Timeliness
7. Fosters Progress and Cooperation with Russia and Others
8. Public and Institutional Acceptance
9. Additional Benefits

After a series of studies, including technical work for a preliminary environmental impact study, DOE arrived at three reasonable disposition alternatives that merited further study to determine which was the best disposition method. Those disposition alternatives are: burning plutonium by using it for reactor fuel; encasing it in other material, thereby immobilizing it and making it inaccessible; and burying it in a deep borehole.

The Programmatic Environmental Impact Statement (PEIS), covering these three options was published in December 1996. In January 1997, DOE announced its Record of Decision (ROD) on plutonium disposition, recommending a dual disposition path: immobilize low-grade plutonium materials and burn high-grade plutonium materials in a reactor. This dual path will provide United States with the basis and flexibility to implement plutonium disposition efforts either multilaterally or bilaterally through negotiations or unilaterally as an example to Russia and other nations. The alternative of burying surplus plutonium in a deep borehole ran into siting and licensing difficulties and was eliminated from consideration, despite the fact that the Lawrence Livermore National Laboratory studies proved it to be technically as feasible as the other two alternatives.

There are two candidate technologies under consideration for immobilization: immobilization in ceramics and immobilization in glass. After the original set of down selections were completed, three variants based on vitrification technology and two variants based on ceramic technology remained under consideration. For the reasons of timing and costs, these five variants have effectively been reduced to two "can-in-canister" variants, in which an inner can containing a plutonium- and neutron-absorber-bearing glass or ceramic is surrounded by a glass containing a radiological barrier, which, in turn is contained in an outer storage canister. A decision between these two is scheduled for September 1997.

## **DISCUSSION**

DOE/MD selected the Lawrence Livermore National Laboratory as Lead Laboratory to study and recommend methods for transformation of Surplus Fissile Materials (primarily plutonium) into long-term immobilized forms meeting environmental, safety, and security objectives; to provide appropriate input to other Disposition Tasks Teams so as to assess technical feasibility of immobilization as a long-term disposition option; and describe infrastructures required to conduct disposition of Surplus Fissile Materials. Support laboratories include Savannah River Technology Center, Argonne National Laboratory, and Pacific Northwest National Laboratory. Several U.S. universities and private industries are also partners, as are several other nations (including Australia, the United Kingdom, France, and Russia) with relevant interests and experience in immobilization.

### **The Spent Fuel Standard**

Because most nations and even some well funded subnational groups are technically capable of remaking surplus plutonium into crude, but politically effective, nuclear weapons, the ideal disposition method would be one that totally eliminates weapons-usable plutonium from the face of the Earth. But

developing and deploying the technology to accomplish this will take more time than the world can afford. If a disposition method is not available within a reasonable timeframe, the growing volume of surplus plutonium will make proliferation easier and render arms reduction agreements meaningless.

Because total elimination is not a practical objective, the U. S. NAS study, commissioned by DOE, proposed the next best thing to manage this "clear and present danger:" minimized accessibility, which is comparable to the accessibility of the plutonium found in much larger and growing stockpile of spent commercial reactor fuel. The "spent fuel standard" is a reasonable goal because the technology to accomplish it appears achievable within 10 years and implementation can be completed within 25 years. It is also a practical goal because it would not require disposition action for spent fuel plutonium, which comprises the larger part of the world's supply of weapons-usable plutonium.

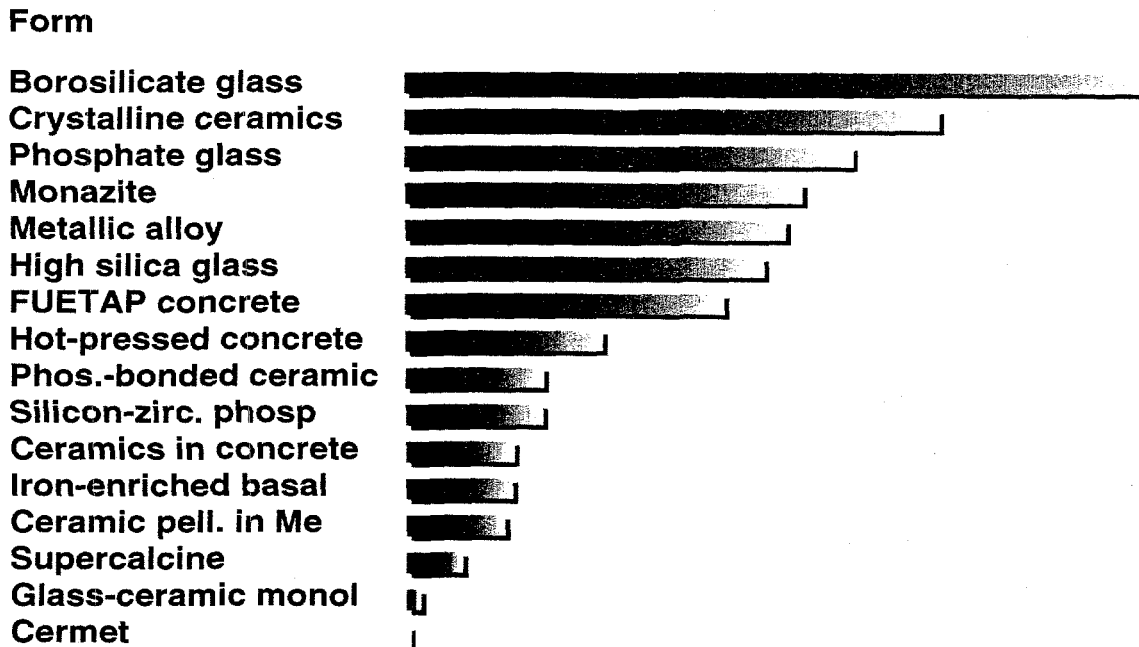
### **Selecting Immobilization Forms**

Because extensive information about stabilizing radioactive materials by embedding them into another material has been published, a literature search was the logical first step taken for the immobilization studies. That search identified 72 forms considered previously for immobilizing radioactive waste. The Immobilization Task Team grouped these forms into families (e.g., calcine, cementitious, ceramic, glasses, and metallic) with common chemical and physical characteristics and then pared them down to a list of 45 by eliminating redundancies among them (e.g., different shape or geometry). Next, the 45 forms were subjected to a formal, two-step screening process to derive top candidates for comprehensive technical evaluation. In the first step, the forms were eliminated if they could not comply with a list of regulatory requirements. Sixteen forms survived for the second step, which used decision analysis principles to quantify the potential of each form to immobilize radioactive waste. These forms were also measured for their desirable attributes using a list of attributes designated and weighted by the researchers. Using these numerical values, they were ranked as shown in Table 1. The two top-ranking forms were borosilicate glass and Synroc (synthetic rock), a ceramic material developed by Australian scientists from the Australian National Science and Technology Organisation and U.S. scientists from Lawrence Livermore and Savannah River.

**Immobilization Processing Options.** The glass and ceramic forms were evaluated in five variations of the immobilization process to look at various permutations of forms, radiological barrier concepts, and facilities in which the work could be done. As in all other disposition methods, the plutonium must first undergo a front-end process that converts it into an oxide, and then a neutron absorber must be mixed with it for criticality control. The five base-case process variations are given in Table 2 and discussed below.



Table 1. Ranking of forms according to weightings and utility curves.



### Three Vitrification (Glass) Variations

Variation 1: Internal Radiation Barrier, New Facility. In this two-stage process, plutonium oxide would react with glass frit containing a neutron absorber to prepare a plutonium-neutron-absorber-glass frit (Figure 2a). First, 4 kilograms or less of plutonium as plutonium oxide would be combined with neutron absorber and glass frit to form plutonium-glass frit. The second step blends batches of 50 kilograms or less of plutonium as plutonium-glass frit with additional neutron-absorber-containing glass frit and  $^{137}\text{Cs}$ , where the cesium would be used as a radiological barrier. The resulting molten glass product is poured into a canister (Figure 2b), welded shut, decontaminated, and stored until permanent disposal in a high-level waste repository.

Variation 2: Internal Radiation Barrier, New and Modified Facility. This two-stage process is similar to Variation 1 (Figure 3). The first-stage melt of plutonium oxide and borosilicate frit (containing a neutron absorber) would be made in an existing facility at Savannah River, and the second-stage melt, which incorporates the cesium radiological barrier, would be done at a new melter to be built next to Savannah River's Defense Waste Processing Facility. The high-level waste fission product  $^{137}\text{Cs}$  would come from the Savannah River tank farms.

Table 2. Immobilization Variants

Immobilization Alternative	Variants	Description
Vitrification	Greenfield	<ul style="list-style-type: none"> <li>• Combined plutonium processing and glass melter facility</li> <li>• A two step vitrification process</li> <li>• Plutonium immobilized in borosilicate glass with <math>^{137}\text{Cs}</math> radiation barrier. <math>^{137}\text{Cs}</math> from <math>^{137}\text{CsCl}</math> capsules at Hanford used as radiation source.</li> </ul>
	Can-in-Canister	<ul style="list-style-type: none"> <li>• Existing facility on DOE site used for plutonium conversion and glass melter facility</li> <li>• Plutonium immobilized in glass in small cans; cans placed into DWPF canisters with HLW glass as radiation barrier</li> <li>• Canister filling done at DWPF</li> </ul>
	Adjunct Melter	<ul style="list-style-type: none"> <li>• A two step vitrification process</li> <li>• Plutonium first dissolved in glass frit in existing plutonium processing facility on DOE site</li> <li>• New adjunct melter adjacent to DWPF as second stage melter.</li> <li>• <math>^{137}\text{Cs}</math> from HLW supernate used as radiation source</li> </ul>
Ceramic	Greenfield	<ul style="list-style-type: none"> <li>• Combined Plutonium processing and ceramic immobilization facility</li> <li>• Plutonium immobilized in ceramic matrix with <math>^{137}\text{Cs}</math> from Hanford <math>^{137}\text{CsCl}</math> capsules as radiation barrier</li> <li>• Ceramic forms placed in DWPF-type canister and backfilled with <math>\text{TiO}_2</math></li> </ul>
	Can-in-Canister	<p>Existing facility on DOE site used for plutonium conversion and ceramic immobilization facility</p> <ul style="list-style-type: none"> <li>• Plutonium immobilized in ceramic matrix , packaged in small cans; cans placed into DWPF canisters with HLW glass used as radiation barrier</li> <li>• Canister filling done at DWPF</li> </ul>

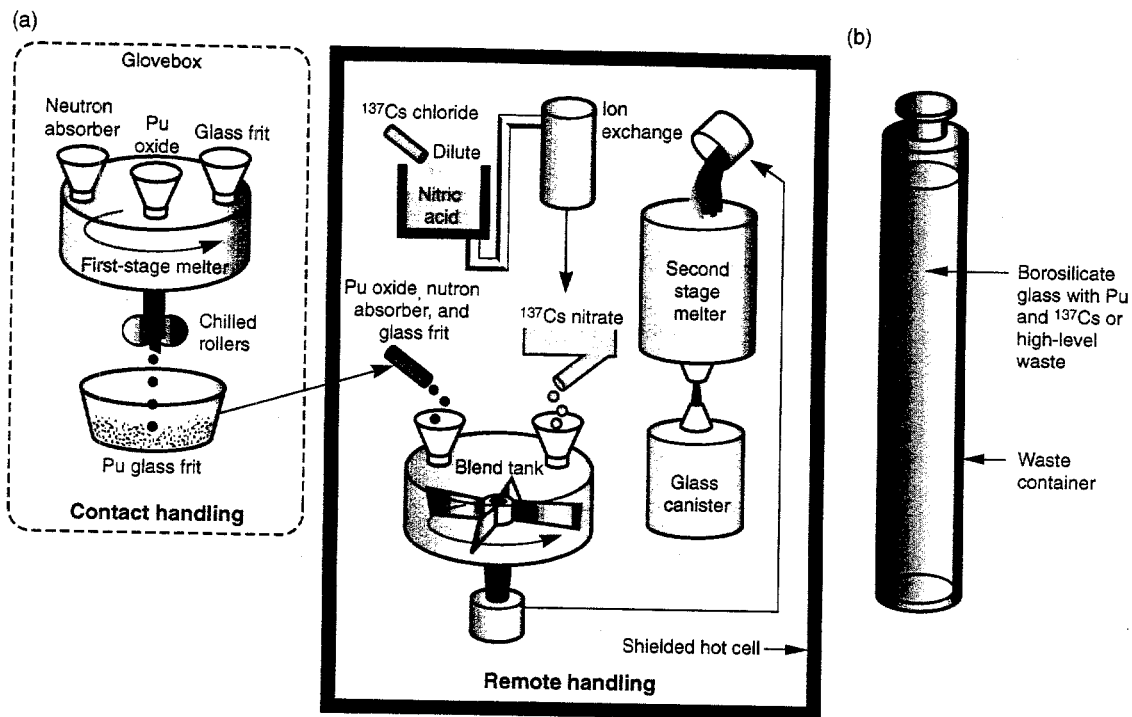


Figure 2. (a) Variation 1 is a two-stage vitrification process in which plutonium oxide is mixed with glass frit and a neutron absorber. (b) The resulting molten glass product is poured into a canister, welded shut, decontaminated, and stored to await permanent disposal.

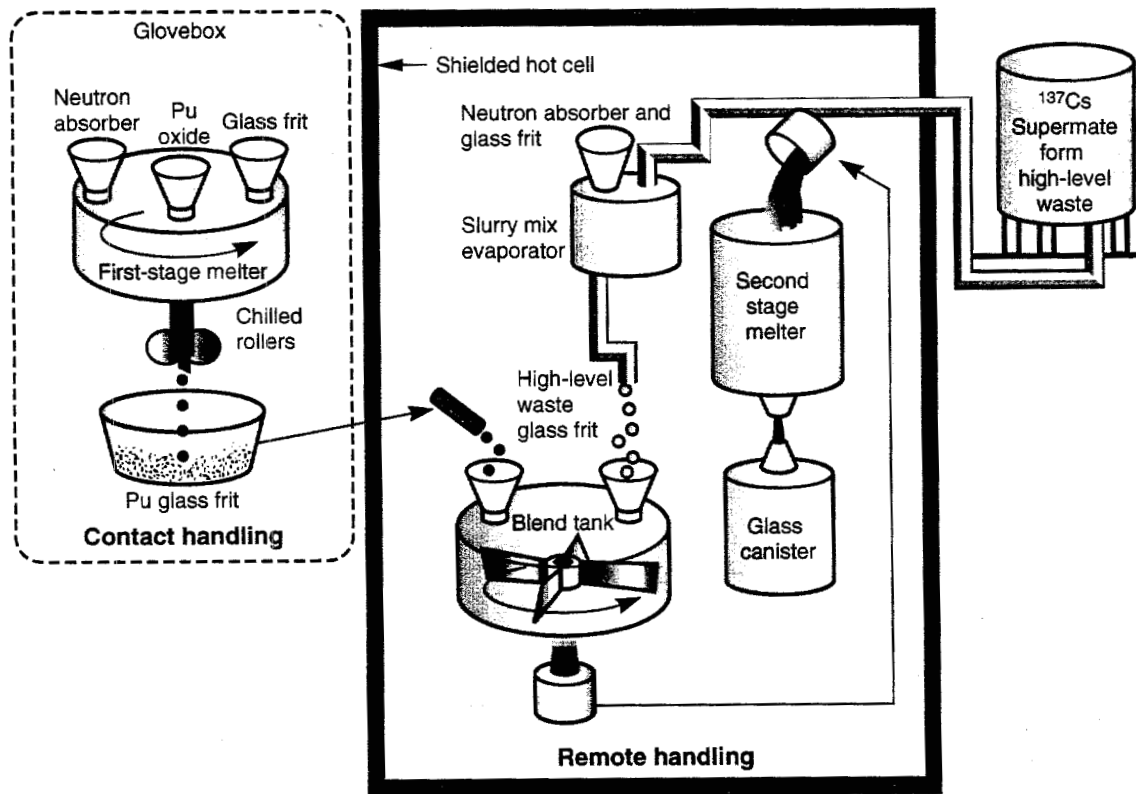


Figure 3. Variation 2 is a vitrification process similar to Variation 1 and like it, produces canistered waste with an internal radiation barrier (Figure 3b). The waste for this variation comes from DOE's Savannah River Defense Waste Processing Facility.

Variation 3: External Radiation Barrier, New Facility. This is a "can-in-canister" concept in which plutonium is immobilized in borosilicate glass, containing a neutron absorber, before being poured into cans, which would in turn be placed in canisters into which molten high-level waste glass would be poured (Figure 4). The high-level-waste glass comes from the Defense Waste Processing Facility at Savannah River.

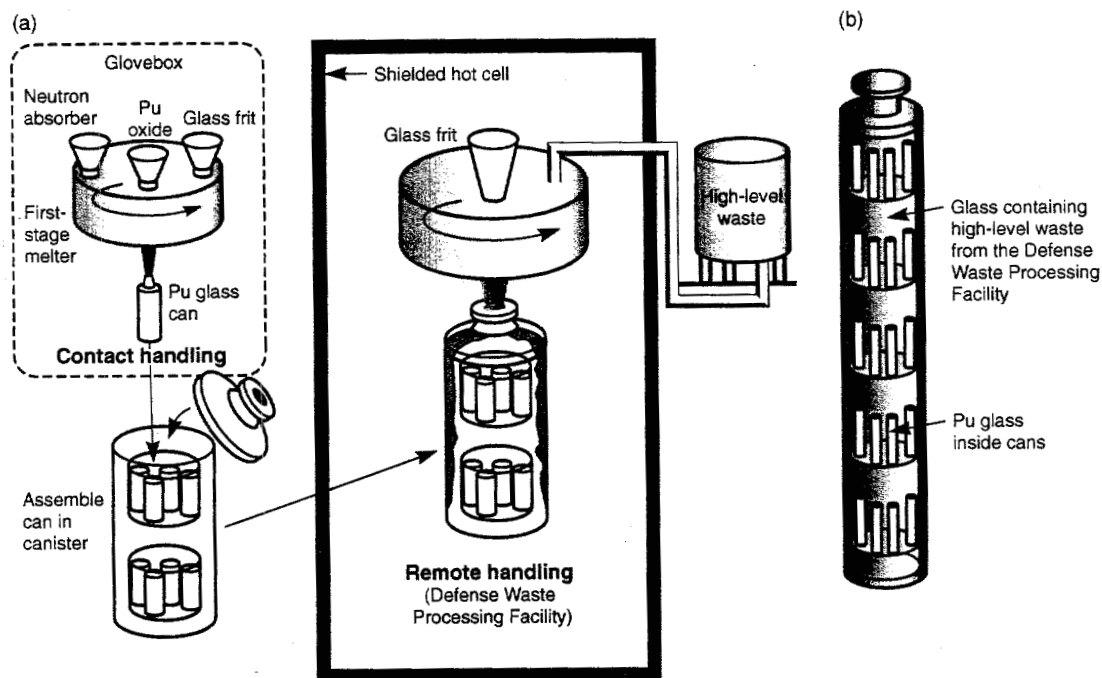


Figure 4. (a) Vitrification Variation 3 is a "can-in-canister" concept in which plutonium immobilized in borosilicate glass is poured into a can, which is then placed in (b) canisters into which molten high-level waste glass from the Defense Waste Processing Facility at Savannah River is poured. The outer canister provides an external radiation barrier.

### Two Ceramic Variations

Variation 4: Internal Radiation Barrier, Modified Facility. Plutonium oxide first would be converted to plutonium nitrate and then blended with mineral-forming oxides (ceramic precursors), a neutron absorber, and a titanate that contains cesium. The mixture would be calcined (heated but not fused), loaded into bellows, and hot pressed into a dense form (Figure 5). Twenty of these forms would be loaded into a canister and packed with titanium oxide granules. The canisters are stored until they could be sent to a high-level waste repository.

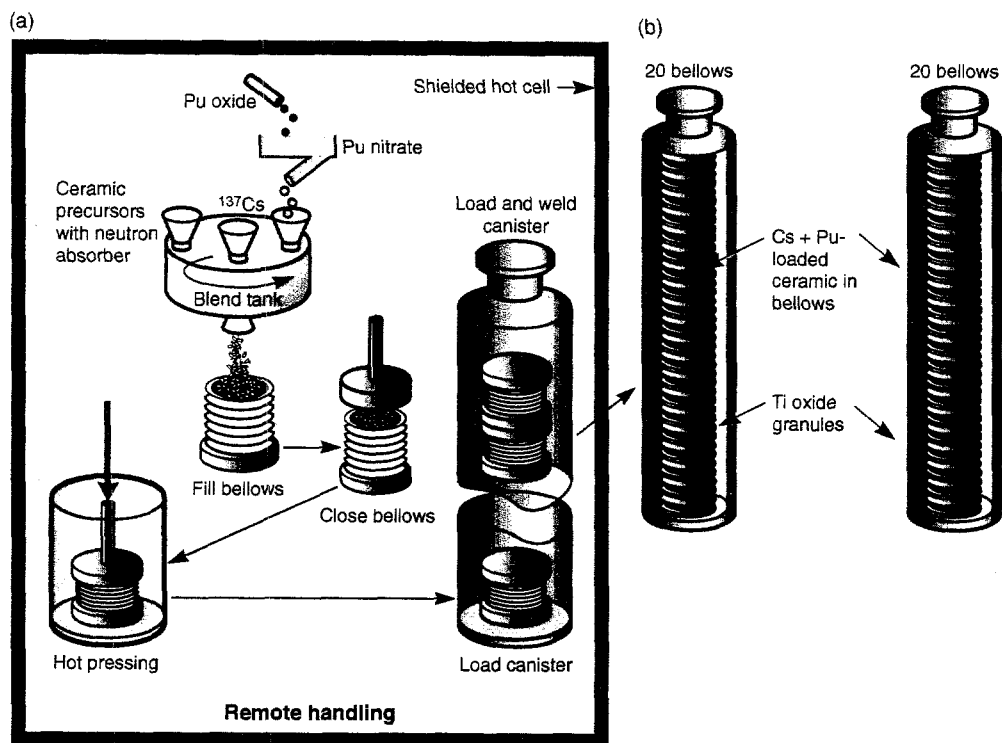


Figure 5. (a) Variation 4 is a ceramic process using an internal radiation barrier. Plutonium oxide is converted to plutonium nitrate and then blended with mineral-forming oxides (ceramic precursors), a neutron absorber, and a titanate containing cesium. The mixture is heated, loaded into bellows, and hot pressed into a dense form. (b) Twenty of these forms will be loaded into a canister, packed with titanium oxide granules, and sent, ultimately, to a permanent high-level waste repository.

**Variation 5: External Radiation Barrier, New Facility.** This is a can-in-canister approach similar to vitrification Variation 3. The ceramic form would be made by blending plutonium oxide with ceramic precursor materials and a neutron absorber. The mixture would be calcined, cold pressed, and sintered (heated but not melted) into a dense form that would be loaded into small cans. The small cans would be put inside a storage canister, where they would be surrounded by glass made with high-level waste (Figure 6).

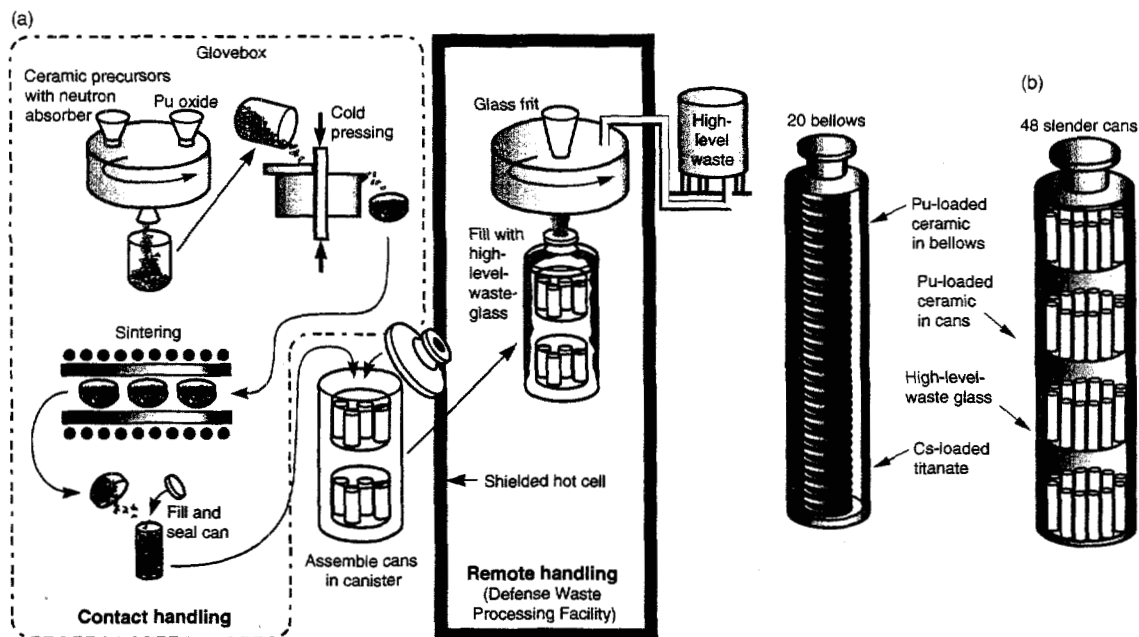


Figure 6. (a) Variation 5 is another ceramic process, but it involves a "can-in-canister" (or external radiation barrier) approach like Variation 3. The ceramic form is made by blending plutonium oxide with ceramic precursors and a neutron absorber. The mixture is heated, cold pressed, and sintered (heated but not melted) into a dense form. (b) It is then loaded into small cans, which are put inside a storage canister and surrounded by glass made with high-level waste.

**Assumptions.** The following assumptions apply for the immobilization alternatives:

- The operational campaign for the immobilization facility will take no more than 10 years to complete.
- The nominal feed of plutonium to the facility is 50 tonnes. Nominal throughput is therefore 25 kg plutonium per day for 200 days of operations per year for 10 years.
- Design for criticality safety will meet applicable DOE Orders and available NRC regulator guides. Criticality is prevented by using batch mass control or equipment geometry as the preferred methods in the designs. The use of appropriate neutron absorbers (e.g., gadolinium, samarium, hafnium) for long-term criticality control has been assumed.
- The immobilization canister assumed for this study shall not exceed 0.6 meter in diameter by 3.0 meters long cylindrical canister.
- The immobilized plutonium package will contain an added radiation field to increase proliferation resistance. The gamma radiation field will be greater than 100 R/hr at 1.0 meter from the package surface 30 years after initial fabrication.

**Technical Approach.** Each immobilization variant was defined for analysis as the beginning-to-end set of operations (e.g., from surplus plutonium to geologic disposal) necessary to address all of the surplus weapons-usable plutonium. We defined and developed the network of operations that is necessary to accomplish the immobilization of materials at a much greater level of detail than was used for either the Screening Report or the NAS Report. The following information was assembled for each of the immobilization variants analyzed:

- Block flow diagrams describing process steps for all operations.
- Lists of major equipment and facilities to accomplish each immobilization function.
- Mass balance and rate data for unit operations and facilities.
- Sketches of equipment layouts and plot plans.
- Reviews of regulatory and operational considerations for facilities.
- Estimates of facility sizes, personnel requirements, and facility infrastructure requirements.
- Identification of balance of plant requirements.



This defined the immobilization variants in sufficient detail to permit technical assessments to be performed, and allowed the analysis of the variants with respect to technical, cost, and schedule criteria. The team also performed necessary experimental and development work required to enhance the knowledge base of immobilization, such as:

- Engineering scale fabrication of ceramic waste forms with plutonium.
- Full-scale "cold" (i.e., without any radionuclides) demonstration of the can-in-canister concept.

### **Record of Decision**

In the recently published Programmatic Environmental Impact Statement (PEIS) and Record of Decision (ROD) for the Storage and Disposition of Weapons-Usable Fissile Materials, DOE announced its decision to pursue two alternative technologies for the disposition of weapons-usable plutonium: (1) irradiation of plutonium as mixed-oxide fuel in existing power reactors, and (2) immobilization of plutonium into large solid forms containing fission products to provide a radiation barrier. The immobilization alternative involves the fixation of surplus weapons-usable plutonium in a stable solid form that is nuclear criticality-safe, proliferation resistant, and environmentally acceptable for long-term disposal in a geologic repository. The Department currently intends to immobilize at least the impure plutonium materials contained in surplus weapon material inventories and in unirradiated plutonium fuels.

### **RD&D Strategy**

The principle driver of the immobilization program is timeliness - specifically, to have a can-in-canister immobilization capability available as early as 2004-2005. The RD&D program described in this plan has been tailored to meet the timeliness and other programmatic objectives with a reasonable level of project risk.

Preliminary R&D completed for the glass and ceramic plutonium forms indicate that the overall "dry" process flowsheets are very similar for the two forms except for the actual technique used to fabricate the plutonium form. Generally, the processes and equipment for first-step immobilization for both glass and ceramic are similar to those used in MOX fuel manufacturing and in the glass manufacturing industry. Thus, the equipment components needed for immobilization can be adapted from existing industrial applications.

Following laboratory-scale development of the immobilization forms in FY 1997, process development and equipment systems testing will be performed and verified at plant scale (i.e., batch size but not throughput) with plutonium using pre-prototypic equipment and subsystems in glove box facilities at LLNL. A fully integrated pilot plant for cold testing of critical integrated prototypic equipment systems would be constructed and the equipments and systems proven prior to plant start-up.

## **R&D Plan**

The ultimate goal of the Immobilization program is to develop, construct, and operate facilities that will immobilize the US surplus plutonium materials with HLW during the next 15 to 20 years. Deployment of the can-in-canister capability should occur by 2004-2005. In support of this ultimate goal, the Research, Development, and Demonstration (RD&D) program has two principle objectives: to establish the technical basis for the design and construction of the US capability to immobilize excess weapons usable plutonium in a suitable and cost effective manner, and to support the United States in influencing Russia to take reciprocal actions to ensure disposition of its surplus plutonium.

Specific RD&D objectives include:

1. Develop an immobilization-waste form that meets nonproliferation objectives for plutonium disposition; has high durability under repository conditions and meets repository acceptance standards; and effectively incorporates desired concentrations of plutonium, uranium, neutron absorbers, and expected impurities.
2. Develop necessary and sufficient technical data on the immobilization processes and processing equipment to support the design of a cost effective immobilization plant that meets ES&H and S&S requirements; has robust processing parameters to ensure quality products; and is sufficiently flexible to accommodate the variable feed materials.
3. Demonstrate the completely integrated process for the can-in-canister technology in a cold (nonradioactive) pilot line at the immobilization sites as a final demonstration of the fully integrate process, to facilitate technology transfer, procedure development, and operator training.
4. Provide an efficient and effective technology transfer from the multiple laboratory development effort to the production plant.

**Plutonium Conversion.** The objective of plutonium material conversion (front end) is to provide the immobilization stage with a blended oxide feed having a leveled plutonium, uranium, and impurity concentration and to levelize both plutonium and uranium isotopic concentrations.

Plutonium material conversion contains the following processing functions: fuel decladding, oxide fuel size reduction, alloy/metal conversion to oxide by hydride/oxidation (HYDOX, Figure 7), the leaching/washing of halides from certain oxide materials, and the coarse blending of the oxides from the various feed streams.

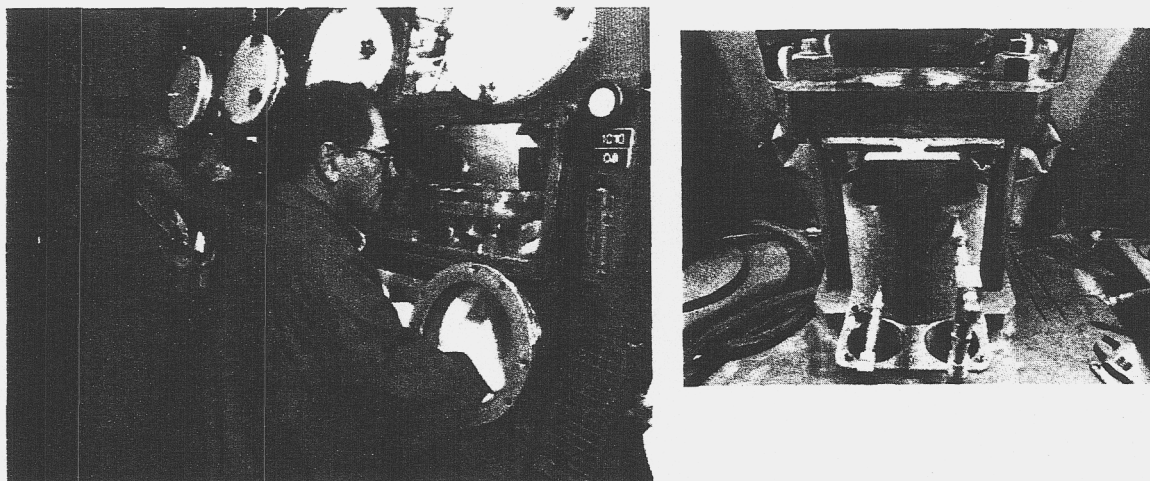


Figure 7. To be suitable for most disposition methods, the plutonium from weapons must first be processed into plutonium oxide. Various hydride/oxidation (HYDOX) methods are used to process the plutonium from weapon pits. The prototype HYDOX furnace design originated and was assembled at Livermore and has been used to test various HYDOX process options

**Immobilization Form Development (Formulation).** Formulation involves the development of an optimum ceramic or glass matrix to incorporate plutonium at the design concentration together with neutron absorbers and the other chemical constituents expected to be present in the surplus feed materials. The chemical and physical properties of the plutonium immobilization forms must be indicative of a solid material which is suitably stable in a geologic repository environment and which can also be produced readily in a production process. Optimization will involve a balance between form performance properties and ease/cost of processing.

In FY97, work will focus on the parallel development of ceramic and glass plutonium forms leading to the selection in September 1997 of a single form for further development and qualification for the immobilization plant and repository. Following form selection, a baseline composition will be selected (April 1998) and fine tuned into a final composition of the selected immobilization form (December 1998) for process prototyping and repository qualification activities. Formulation activities during FY99 and beyond will support the important task of qualifying the plutonium immobilization form for the repository.

**Immobilization Form Characterization and Repository Acceptance.** The plutonium immobilization form must be qualified for acceptance in a federal geologic repository. The main issue with the plutonium form is the possibility that the fissionable nuclides  $^{239}\text{Pu}$  and  $^{235}\text{U}$  will separate from the neutron absorbers over geologic time periods. For this purpose, testing techniques have been developed and are being used to measure the long-term stability of the plutonium immobilization forms. In addition, data are being generated and models are being developed to assess the potential chemical reactions and geologic transport characteristics of the immobilization form constituents.

**Immobilization Process Development.** This program element develops the processing technology and equipment used to convert the oxide feed material produced by the plutonium conversion subsystem into the final solid plutonium form. At this time the point design for both the ceramic and glass processes are based upon dry flowsheets. A major portion of both flowsheets share similar unit operations. These operations involve milling, grinding/mixing, and granulation of the material produced by the plutonium conversion process together with either ceramic precursors or glass frit to produce an acceptable oxide powder for the fabrication step. This oxide powder is then cold pressed and sintered in the ceramic process or fed to a glass melter in the glass process to produce the final immobilized plutonium form.

This consists of two major tasks. The first is the development of unit process operations and testing of key equipment at full scale, both using

nonradioactive surrogates and with plutonium, during FY1998-1999 to provide technical data on processing conditions and equipment for plant design. The critical unit operations include: milling, blending, granulation, glass melting, or ceramic forming equipment. In addition, development associated with a fully integrated nonradioactive process line will be performed to determine the following system integration characteristics: the behavior of powder material transport between unit operations, dust containment, and material holdup. These important system integration characteristics and parameters are also required as input to plant design.

Development work with nonradioactive surrogates will be performed at both LLNL and SRS in a complimentary manner. The plutonium process development activities will be performed by LLNL and WSRC personnel in the existing plutonium glove box facilities at LLNL. During FY1998-1999, plutonium process developments work, jointly carried out by LLNL and SRTC personnel will provide valuable information for the pilot plant, again using nonradioactive surrogates, planned for SRS in late FY1999.

### **Plant Project - Design, Construction, and Activation**

The plant design and construction element follows the prescribed steps of conceptual design, detailed design (Title I and Title II), and construction. There is a very tight time window for the development program to deliver the necessary technical data (design criteria and requirements, process conditions, equipment specifications, etc.) to the project design activity. In parallel with the plant design, safety documentation must be developed and regulatory approvals are obtained for plant activation and operation. Nonradioactive activation of the plant is scheduled at the beginning of FY2004. One year of nonradioactive operation is allocated to conduct the Operational Readiness Review (ORR) for start of plutonium operations, and to use the integrated plant equipment for final qualification of the plutonium immobilization process and form.

### **Integration and Support Activities**

The integration and support program activities which are important to the overall program but which either span across the other program elements or do not logically fall within one of the other elements. These activities include work to enhance the proliferation resistance of the immobilization form, and mutual interactions with the Russians on plutonium disposition.

The goal of the proliferation resistance task is to develop and demonstrate concepts to enhance the robustness of the can-in-canister system against unauthorized attempts to physically extract the plutonium cans from the

canister. These concepts include: melting cans, welding the can structure to the canister, internal armor, etc. In FY98, the most promising concepts analyzed in FY97 will be experimentally tested cold (nonradioactive) by fabricating canisters containing the selected internal can configuration, and pouring simulated HLW glass using melters available either at PNNL or Clemson University.